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(54) Adhesive Composition for Laminate

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## Specification

1. Title of the Invention Adhesive Composition for Laminate

2. Extent of Patent Claims

What Is Claimed Is:

An adhesive composition for an electron beam hardening laminate consisting of a compound with an ethylenic unsaturated bond and a linear saturated polyester resin composing between 5 and 60% by weight of the total.

## 3. DETAILED DESCRIPTION OF THE INVENTION

The present invention relates to an adhesive composition for an electron beam hardening laminate. In further detail, the present invention relates to an adhesive composition for an electron beam hardening laminate that can provide an adhesive layer that is flexible and has excellent adhesive force.

Various types of lamination films are being used, primarily as packaging materials. Methods to produce these lamination films are already known, including dry lamination, extrusion lamination, and wet lamination, and the like. Adhesives are used for this lamination, and for instance polyurethane adhesive, polyacrylic ester adhesive, epoxy resin adhesive, vinyl chloride/vinyl acetate copolymer adhesive, and isocyanate adhesive, and the like, are used. However, these adhesives are dissolved and used in organic solvents such as ethyl acetate, methyl ethyl ketone, toluene, xylene, or hexane, or the like, so during the adhesion operation or during drying, large quantities of organic solvents are dispersed into the atmosphere, which not only has the problem of significantly degrading the working environment, but also is very uneconomical from a thermal perspective. Furthermore, because trace amounts of organic solvent cannot be prevented from remaining between the laminate films, the use in applications where odors are undesirable or in packaging for materials where odors can easily be absorbed, and even if used, product value will be degraded by the odor of the residual solvent. Furthermore, the use of large amounts of organic solvent is not preferable from the viewpoint of consuming natural resources.

In contrast, an electron beam hardening adhesion method which uses a solvent-free adhesive not only eliminates the problems with a conventional laminate method which uses a solvent diluted adhesive as described above, but also provides advantages such as the line speed can be increased so productivity is increased, a long drying furnace is not necessary, equipment floor space is reduced, production control is simplified with momentary start up and stopping, and material losses during start up and stopping are minimized, so broad use is anticipated, primarily for lamination films using packaging material. However, adhesives which use this method are expected to harden completely, so there are disadvantages in that the flexibility of the hardened layer obtained is low, and the adhesive strength is still insufficient.

The present invention is a new composition that improves the flexibility and adhesion of adhesives for conventional electron beam hardening laminates, and consists of a compound with an ethylenic unsaturated bond and a linear saturated polyester resin composing between 5 and 60% by weight of the total. If the amount is less than 5%, the flexibility of the hardened adhesive will be poor, but if the amount exceeds 60%, the viscosity of the composition will be high, and the painting process will be difficult.

The linear saturated polyester resin of the present invention consists of a diacid base residue and a diol residue, and resin which has a molecular weight between 10,000 and 30,000, a limiting viscosity between 0.5 and 1.0, and a glass transition temperature of 10°C or lower is suitable for the present invention. Examples of the dibasic acid component include aromatic dicarboxylic acids such as terephthalic acid, isophthalic acid, 1, 2-bis(P-carboxyphenoxy) ethane, 2, 2-bis(P-carboxyphenyl) propane, and aliphatic carboxylic acids such as dicarboxylic acid, succinic acid, adipic acid, and subacic acid and the like. Examples of the diol component include aliphatic, aromatic, and alicyclic diols such as ethylene glycol, propylene glycol, tetramethylene glycol, pentamethylene glycol, 2, 2-dimethyltrimethylene glycol, hexamethylene glycol, P-xylene glycol, and cyclohexane dimethanol. Furthermore, the diol component may also be a block copolymer polyester which has as polymer components aliphatic polyester glycols such as polyethylene glycol, polypropylene glycol, polytetramethylene glycol, polyethers such as the copolymers of ethylene oxide and propylene oxide, polyneopentyl acetate, and polyneopentyl adipate, and the like.

Examples of the compound with an ethylenic unsaturated bond of the present invention include polyester (meth)acrylate, polyurethane (meth)acrylate, polyepoxy (meth)acrylate, polyether (meth)acrylate, and other prepolymers; styrene, α-styrene, vinyl acetate, and other olefinic monomers; methyl acrylate, 2-ethylhexyl acrylate, 2-hydroxyethyl acrylate, methoxyethyl acrylate, butoxyethyl acrylate, butyl acrylate, methoxybutyl acrylate, cyanoacrylate, cyanoethyl acrylate, phenyl acrylate, and other acrylate esters; methyl methacrylate, propyl methacrylate, methoxyethyl methacrylate, ethoxymethyl methacrylate, phenyl methacrylate, ethyl methacrylate, lauryl methacrylate, and other methacrylate esters; acrylamide, methacrylamide, N-N-dimethylacrylamide, N-N diisopropylamide, N,N-didecylacrylamide, N,N-dimethyl methacrylamide, N,N- diethylmethacrylamide and other unsaturated carboxylamide and derivatives thereof; 2-(N,N-dimethylamine) ethyl acrylate, 2-(N,N-dimethylamino) ethyl acrylate, 2-(N,N-dibenzylamino) ethyl acrylate, (N,N-dimethylamino) methyl methacrylate, 2-(N,N-diethylamino) propyl acrylate and other similar compounds; ethylene glycol diacrylate, propylene glycol diacrylate, neopentyl glycol diacrylate, 1,6-hexanediol diacrylate, diethylene glycol diacrylate, triethylene glycol diacrylate, dipropylene glycol diacrylate, ethylene glycol dimethacrylate, propylene glycol dimethacrylate, diethylene glycol dimethacrylate, and other by functional monomers, trimethylol propane triacrylate and other trifunctional monomers or similar compounds, and these compounds may be used individually or two or more may be used as a blend.

The adhesive composition of the present invention is prepared by blending and kneading using standard methods while heating the aforementioned components if necessary to make an adhesive with a viscosity in a range between 100 and 800 cp. Note, if necessary, plasticizers or polymerization inhibitors, or the like, may be added to the adhesive composition of the present invention.

The composition of the present invention is used for adhesion between similar or different types of films such as paper, plastic, or metal, and the like. Examples of plastic films include stretch or non-stretch films of various resins such as cellophane, low density, medium density, or high density polyethylene, polypropylene, polyethylene terephthalate, polyamide, polycarbonate, polyacetate, polyvinylchloride, polyvinylidenechloride, polyvinyl alcohol, ethylene-vinyl alcohol copolymer, ethylene-vinyl acetate copolymer, ethylene-α- olefin copolymer, ionomer resin, and poly-1, 2-butadiene, as well as metal vapor deposition films such as aluminum, or the like, or paint films such as polyvinylidene chloride, vinyl chloride-vinyl acetate copolymer, or hardened [illegible] formed on these plastic films. If necessary, these films may be surfaced treated with a Corona treatment, or the like, prior to application of the adhesive.

In order to obtain a laminate film using the adhesive composition of the present invention, all or any part of a base material film is coated with the composition of the present invention at a rate of between 0.2 and 20 $g/m^2$, preferably between 0.4 and 10 $g/m^2$, using a gravure coating method, roll coating method, or the flexo-reverse method, or the like, and then applying a different film (either the same or a different type than the base material film), or extruding another film base material and then laminating multiple layers together, after which the adhesive composition is hardened using electron beam irradiation. Laminate films with three or more layers can be obtained by a method where the aforementioned method is repeated using a multiple layer film as a base material, or a method where 3 or more films to which the adhesive of the present invention has been applied are overlaided and multiple layers of adhesive are hardened by a single electron beam irradiation.

The electron beam used to harden the adhesive is an electronic beam with energy in the range between 50 to 1000 KeV, preferably between 100 and 300 KeV, irradiated from one of various types of electron beam accelerators including a Cockcroft-Walton accelerator, Van de Graaff accelerator, resonance transformer accelerator, insulated core transformer accelerator, linear accelerator, dynamitron accelerator, or high-frequency accelerator, or the like. In order to irradiate the adhesive layer with an electron beam, either a method where a spot beam travels at high speed or a method where a continuous curtain shaped beam is continuously irradiated from a linear filament is used. Normally surface irradiation is performed by continuously moving the object that is subject to irradiation.

The adhesive composition of the present invention is basically a solvent free composition. Therefore, a laminate film made odor-free by completely hardening using electron beam irradiation in this manner can be extremely useful as a packaging material for various types of food and drugs.

The present invention will be more specifically described below using the following embodiments.

Embodiment 1

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|---|---|
| Pyron 300 (product of Toyo Boshoku)(linear saturated polyester) | 30% by weight |
| Elestra XRA/234 (product of Mitsui Toatsu) (polyester acrylate) | 30% by weight |
| 2-hydroxyethyl acrylate | 40% by weight |

The aforementioned formulation was heated to 125°C to completely dissolve the linear saturated polyester which was a solid, in order to obtain a uniform liquid composition with a viscosity of 700 cp (measured at 25°C).

This composition was applied to a 12 µm thick biaxially elongated polyethylene terephthalate film which had been corona treated, at a rate of 40 g/m$^2$ using a gravure roller, and a 50 µm thick polyethylene film which had been corona treated similar to as described above was overlaid. This combined film (width 450 mm) was rolled at a rate of 60 m/minute in a nitrogen environment while a 150 keV, 5 mA curtain-shaped electron beam was irradiated from the biaxially elongated polyethylene terephthalate film side at an intensity of 1.0 Mrad from a low energy electron beam accelerator (product of Energy Science, Electrocurtain CB20d50/30), in order to harden the adhesive. The adhesive strength (tensile speed 300 mm/min, tensile angle T shaped) of the laminate film obtained was 830 g/15 mm.

Furthermore, a 60 µm polypropylene film was used in place of the polyethylene film of the aforementioned embodiment, and when a test was performed using the same adhesive, the adhesive strength obtained was 760 g/15 mm.

Embodiment 2

| | |
|---|---|
| Pyron 300 (product of Toyo Boshoku)(linear saturated polyester) | 45% by weight |
| Aronix 6100 (product of Toa Gohsei) (oligoester acrylate) | 30% by weight |
| 2- hydroxyethyl acrylate | 25% by weight |

The above formulation was heated to 125°C to completely dissolve the linear saturated polyester which was a solid, in order to obtain a uniform composition with a viscosity of 580 cp (measured at 25°C ).

The same adhesion test as Embodiment 1 was performed using this composition. The adhesive strength of the laminate film obtained was 810 g/15 mm between a biaxially elongated polyethylene terephthalate film and a polyethylene film, and was 790 g/15 mm between a biaxially elongated polyethylene terephthalate film and a polypropylene film.

Agent Patent Attorney ONISHI, Atsumi

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Procedural Corrections

August 8, 1980

Patent Office Commissioner KAWAHARA, Yoshio

1. Presentation of Case
   1980 No. 83,086

2. Title of the Invention
   Adhesive Composition for Laminate

3. Revisor

| | |
|---|---|
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7. Subject of Revision
   Section of "Detailed Description of the Invention" in the Specification

8. Content of Revisions

(1) A "," was deleted in line 5 of page 4 of the specification.

(2) [Japanese spelling correction] was made on the 2nd line from the bottom of page 8.